IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant(s): Donald R. Huffman, et al. **Examiner: Hendrickson**

Serial No.: 07/580,246 **Art Unit: 1754**

Filed: September, 10th 1990 **Docket: 7913Z**

For: NEW FORM OF CARBON

Assistant Commissioner for Patents
Washington, DC 20231

DECLARATION OF Raouf O. Loutfy

I, Raouf Loutfy, hereby declare:

1. I reside at 6507 N. Ventana Canyon Drive, Tucson, Arizona, USA.

2. I earned a B.Sc. degree in 1964 in Applied Chemistry from Cairo University, a M. Sc. in 1966 in Solid State Sciences from the American University, a Ph.D in 1971 in Physical Chemistry from University of Western Ontario, and a Diploma in Business Administration in 1976 from McGill University.

3. Between 1977 and 1981 I was employed as a group leader at Argonne National Laboratory in the Chemical Engineering Division.

4. Between 1981 and 1985 I was employed as Research Advisor of ARCO Metals/ARCO Chemical developing advanced material technologies.

5. Between 1985 to present I have been employed as president of MER Corporation, and since 1990 I have been involved in the commercial scale-up of fullerenes, and in developing applications for fullerenes.

6. I have published many articles, two books, and contributed a chapter in Encyclopedia of Technology on the fullerene production, and on applications of fullerenes. I have been an invited speaker to many technical and investment conferences as an expert in the technology and commercialization of fullerenes. I have received the prestigious Tibbetts award in 2001 from the Small Business Administration (SBA) for the commercialization of fullerenes. For the convenience of the patent and trademark office, I have attached hereto as Exhibit 1 my curriculum vitae, which describes my credentials and demonstrates my expertise in the area of fullerenes.

7. I am intimately familiar with the published literature concerning fullerenes and I am personally involved in the research and development of new methods of production and applications for fullerenes including C_{60}, C_{70} and nanotubes.

8. I am informed by Mark J. Cohen, Esq., the attorney handling the prosecution of the subject United States patent application that a question has arisen concerning the possibility of fullerenes molecules formation in the carbon arc operated under conditions

employed in the paper "Spectroscopy of Matrix-Isolated Carbon Cluster Molecules between 200 and 850 nm Wavelength" published in Surface Science, v.156, pp.814-821 (1985) by W.Kratschmer, N.Sorg and D.Huffman.

9. As set forth above, I have been professionally involved in the field of fullerenes development and manufacture since Dr. Huffman and Kratschmer disclosed their novel process for producing macroscopic quantities, i.e., visible quantities of C_{60} and C_{70}, as disclosed in the subject patent application and in their publication in Nature, 347, No. 6391, pp. 354-358 (1990).

10. I have read and reviewed the subject application, including the pending claims. It is my understanding that the application is directed to, among other things to fullerenes, for example, C_{60}, and C_{70} prepared in macroscopic amounts and the process for making same. It is also my understanding that the claims in the subject applications are directed, among other things to C_{60} and C_{70} in macroscopic amounts. I have also read the paper on "Spectroscopy of Matrix-Isolated Carbon Cluster Molecules between 200 and 850 nm Wavelength" published in Surface Science, v.156, and pp.814-821 (1985) by W.Kratschmer, N.Sorg and D.Huffman.

11. I directed experiments to simulate the experiments of Kratschmer et. al. referenced above. We used an existing Quartz graphite evaporator that we normally use to produce fullerenes and fullerenes soot. The double walled quartz chamber is 6" diameter by 10" tall, with quartz flanges at each end. The end pieces are water-cooled

aluminum with a ferrofluidic rotary feed through at the top and a differentially pumped linear/rotary feed through at the bottom to permit movement of the electrodes. Gasses are admitted through fittings in the upper lid using one or more flowmeters and valves. A feedback controller with proportional solenoid valve in the vacuum line automatically controls pressure. Power is supplied by a 3 kW constant-current DC arc power supply. A graphite rod is installed in the lower electrode (anode), and after evacuation and adjustment to the proper gas pressure, is moved via a stepper motor to contact the graphite upper electrode (cathode). The gap is maintained by adjustment of the stepper motor speed. Gas pressure and flow rate, rod feed rate and current are maintained constant. The voltage is allowed to vary, but remains relatively stable while equilibrium conditions of anode rod consumption are maintained. The wall of the Quartz reactor is normally water-cooled; however, for the present experiments we also liquid nitrogen cooled the reactor wall to condense Argon and entrain and matrix isolate soot in the condensed Argon. A single rod is consumed at a time, and products are recovered for each run. This reactor system very much simulate the apparatus in Figure 1 of the Kratschmer et al referenced paper, the left part of (the soot generating part) is exactly the same except our system is computerized in term of rod feeding rate, and with the nitrogen cooled reactor wall simulating the right part of the apparatus.

12. As a reference, I performed a first experiment that typically produces fullerenes-containing soot. In this experiment a ¼ inch in diameter graphite rod was vaporized, under a 100 Torr Helium pressure, using 100-ampere dc current. The graphite rod was vaporized, and the vapor was condensed on the water-cooled Quartz reactor wall. The

vaporization was performed for 50 minutes using about 17 cm length of the graphite rod and produced 12 gram of soot. The fullerenes were recovered using toluene and the amount of fullerene was determined. The yield of fullerene was about 10%. Accordingly, the total recoverable fullerenes was about 1.2 grams with over 900 mg of C_{60} and over 200 mg of C_{70} and the remaining other fullerenes.

13. I performed a second experiment with the same equipment used in the first experiment, operated under different conditions that simulate Kratschmer et al conditions. The reactor was hard evacuated to about 10^{-06} Torr of Argon pressure, and was liquid nitrogen cooled to allow Argon to condense on the wall of the reactor. In this second experiment a ¼ inch in diameter graphite rod was vaporized, under this hard vacuum condition, using 100-ampere dc current. The graphite rod was vaporized, and the vapor was condensed on the liquid nitrogen-cooled Quartz reactor wall. The vaporization was performed for 5 minutes using about 5-cm length of the graphite rod and produced 3 gram of soot embedded in the condensed Argon. The rate of vaporization was significantly higher than that encountered under the normal condition described above in the first experiment for fullerenes production. The reactor was allowed to reach room temperature and the soot was collected from the wall. This soot was analyzed for fullerenes using standard HPLC procedure (Absolute detection limits are on the order of 50 micrograms per liter, 50 ppb), and there no were fullerenes detected.

14. I further performed a third experiment with the same equipment using water-cooled reactor but under hard vacuum of about 10^{-06} Torr of Argon pressure to determine

whether the effect is due to the hard vacuum or the low temperature at the wall of the reactor. In this experiment a ¼ inch in diameter graphite rod was vaporized, under this hard vacuum condition, using 100-ampere dc current. The graphite rod was vaporized and the vapor was condensed on the water-cooled Quartz reactor wall. The vaporization was performed for 20 minutes using about 17-cm length of the graphite rod and produced 12 gram of soot collected on the wall of the reactor. The rate of vaporization again was significantly higher than that encountered under the normal condition described above in the first experiment for fullerenes production. This soot was analyzed for fullerenes using standard HPLC procedure, and again there were no fullerenes detected. Therefore, the key factor that resulted in the absence of fullerenes in the soot is the low pressure. Accordingly, there is no way that fullerenes would have been produced under the conditions described in the referenced paper.

I further declare that all statements made herein of my own knowledge are true, and all statements made on information and belief are believed to be true. I acknowledge that willful false statements and the like are punishable by fine or imprisonment or both (18 U.S.C. §1001) and may jeopardize the validity of the application or any patent issuing thereon.

July 16 2002
Date

Raouf O. Loutfy
Raouf O. Loutfy, Ph.D

Signed in Tucson, Arizona

Exhibit # 1

Curriculum Vitae of Dr. RAOUF O. LOUTFY

EDUCATION

Diploma	Business Administration, McGill University, 1976
Ph.D.	PhysicalChemistry/Electrohemistry, University of Western Ontario, 1971
M.Sc.	Solid State, American University, 1966
B.Sc.	Applied Chemistry, Cairo University, 1964

PROFESSIONAL EXPERIENCE

1999 – Present COO, FIC Corporation, New York, NY
Responsible for technical direction of this joint venture between MER, Mitsubishi Corporation, and RCT Corporation for the commercialization of Fullerene and Fullerene based materials.

1985 – Present President, MER Corporation, Tucson, Arizona
Responsible for developing advanced programs and technologies, managing research and development group, contract administration and financial responsibilities. Produced SiC whiskers, SiC Fibers, ceramic-ceramic composites, advanced lithium-ion battery technology, low cost bipolar plates for fuel cell. Lead a group to develop scale-up production of fullerenes, and its applications development.

1986 - 1988 President, Keramont Research Corp., Tucson. AZ
Responsible for building the infrastructure (equipment, personnel, and projects) for advanced materials research organization with emphasis on electronic ceramics, aluminum nitride substrates reinforcements (SiC_w, SiC_f and $TiB2_w$) and composites (ceramic-ceramic, and metal-ceramic. and intermetallic ceramic).

1981 - 1985 Research Advisor, ARCO Metals/ARCO Chemical Company, Tucson. AZ
Major responsibility in the development of advanced technology to produce primary light metals and advanced materials, and to provide company-wide support in area of expertise. These efforts result, amongst others, in a commercial plant for the production of high purity alumina.

1977–1981 Group Leader, Chemical Eng. Div., Argonne National Laboratory, IL
The major responsibility, as a group leader of the Electrolytic Technology Research Group, was to develop and implement a plan for the electrochemical technology for energy and resource saving. This was achieved by supporting and conducting research and development to improve industrial processes and identifying and developing new concepts of low energy alternative technologies. A second responsibility is the technical management of contracts in the electrolytic area and contracts for developing batteries for load-leveling applications.

1972-1977 Group Leader, Noranda Research Center, Pointe Claire, Quebec
In charge of developing advanced pyro metallurgical and electrometallurgical processes to improve plant operations and profitability. Those efforts led to the development of DSA anodes for metal winning, and a new oxidant for zinc purification process.

AWARDS

TITLE OF AWARD	DATE	PRESENTED BY	REASON FOR AWARD
Industrial R&D 100	1990	R&D Magazine	Development of SiC Fibers
Corporate Entrepreneur of the year	1990	AIN	Most Number of Contract Won in Arizona
Industrial R&D 100	1991	R&D Magazine	Development of SiC Whisker
Product of the Year	1991	AIN	Fullerenes Production
Industrial R&D 100	1996	R&D Magazine	PDS Powder
Tibbetts Award	1998	SBA	SBIR Commercialization
Tibbetts Award	2001	SBA	Commercialization of Fullerene

PUBLICATIONS AND PATENTS

- 24 U.S. patents (12 have been in the last 10 years)
- 12 patent disclosures
- 90 articles published
- A chapter on Hydrogenated Fullerene in the Encyclopedia of Technology
- 2 books, both on Fullerenes:

  D. Koruga, S. Hameroff, J. Withers, R.O. Loutfy, and M. Sundareshan "Fullerene, C60: History, Physics, Nanobiology, and Nanotechnology" Elsevier Science Publishing Co. New York, NY 1993.

  "Perspectives of Fullerene Nanotechnology", edited by Eiji Osawa, Kluwer Academic Publishers, February 2001.

The specific Chapters are:

1. R.O.Loutfy, A. Moravsky, A. Franco, and E.Veksler "Physical Hydrogen Storage on Nanotubes and Nanocarbon Materials"
2. R.O.Loutfy, S.Katagiri "Fullerene Materials for Lithium-ion Battery Applications"
3. R.O. Loutfy, S. Hossain, A. Moravsky and M. Saleh "Nanotubes as Anode Material for Lithium-ion Batteries"
4. Raouf O. Loutfy and Eugene M. Wexler "ABLATIVE AND FLAME-RETARDANT PROPERTIES OF FULLERENES"
5. R.O. LOUTFYJ. C. WITHERS, AND M. ABDELKADER "Development of Carbon Nanotube - Polymer Composites"
6. Raouf O. Loutfy, J.C.Withers and Stevan T. Dimitrijevic "USE OF FULLERENES AND CARBON NANOTUBES FOR FABRICATION OF EFFICIENT ELECTRON FIELD EMITTERS"
7. Raouf O. Loutfy, Eugene Weksler, and Weijiong Li "UNIQUE FULLERENE-BASED HIGHLY MICROPOROUS CARBONS FOR GAS STORAGE"
8. Raouf O. Loutfy, Timothy P. Lowe, Alexander P. Moravsky, and S. Katagiri "Commercial Production of Fullerenes and Carbon Nanotubes"
9. Raouf O. Loutfy, Eugene Weksler "GAS PHASE HYDROGENATION OF FULLERENES"

10. Raouf O. Loutfy, Eugene Weksler "HYDROGENATION OF ALKALI METAL – DOPED FULLERENES"
11. R.O.Loutfy, and M.Hecht "Aligned Carbon-Nanotubes for Sensor Applications"
12. Raouf O. Loutfy, Eugene Weksler "ADVANCED THERMAL PROTECTION COATING USING FULLERENES"

Other Fullerenes Publications:

S. Seraphin, J.C. Withers, R.O.Loutfy, et al, "TEM Studies of Nanotubes and Graphite Particles", Symposium of the Arizona Fullerene Consortium, November 6, 1992.

S. Seraphin, J.C. Withers, et al, "TEM Study of Carbon Nanotubes Produced by Various Processing Conditions," to be presented a John M. Crowley Symposium, Arizona State University, January 5-8, 1993.

S. Seraphin, J.Jiao, D. Zhou, J.C. Withers, R.O. Loutfy, "Effect of Processing Conditions on the Morphology and Yield of Carbon Nanotubes," *Carbon*, Vol 31, No 5, 685 (1993).

S. Seraphin, J. Jiao, D. Zhou, J.C. Withers, R.O. Loutfy, "Yttrium Carbide in Nanotubes," *Nature*, Vol. 362, April 8, 503 (1993).

J.C. Withers, R.O. Loutfy, K.Y. Donaldson, D.P. Hasselman, "Thermal Diffusivity/Conductivity of Compacts of C_{60} Buckminsterfullerene and a C_{60}/C_{70} Mixture, "*J.Am. Cerm. Soc.*, Vol 76, No 3, 754 (1993).

D. Koruga, J.S. Kustic, M. Trifunovic, S. Jankovic, S. Hameroff, J.C. Withers, R.O. Loutfy, "Imaging Fullerene C_{60} with Atomic Resolution Using a Scanning Tunneling Microscopy", *J. of Full. Science & Technology*, Vol 1, 93 (1993).

R.O. Loutfy, J.C. Withers, "Fullerene & Electrochemical Hydrogen Storage" abstract 2145, Electrochem. Soc. Mtg, Hawaii, (1993).

J.C. Withers, R.O. Loutfy, "Production Process for Fullerenes, Review" Abstract #2127 Electrochem. Soc. Meeting, Hawaii (1993).

T. Yadav, S. Seraphin, D. Zhou, J.C. Withers, R.O. Loutfy, "Catalytic Growth of Buckyonions," in preparation

S. Supapan, D. Zhou, J.Jiao, M. Minke, S. Wang, T. Yadav, J.C. Withers, R.O. Loutfy, "The Effect of Pt, Pd and Ni on the Synthesis of Carbon Clusters," in preparation

J.C. Withers, C. Pan, R.O. Loutfy, "Fullerene Price: How Low Will They Be?" Electrochemical Society Mtg., San Francisco, Abstract 1216, (1994)

Contracts

DOE contract #DE-FG02-92ER81272, "The Development of a Process to Synthesis Tubular Fullerenes", Phase II, July 92

DOE contract #DE-FG02-91ER81095, "Novel C_{60} Electrodes for Advanced Electrochemical Sensors", Phase I, Sept 91.

ARMY contract #DAAH04-93-C-0004, "Novel Materials for Hydrogen Supplies and Storage for Fuel Cells, Phase I, Jan 93.

ARMY contract #DASG60-93-C-0003, "Use of High Energy Lasers for Materials Synthesis", Phase I, Nov 92.

NASA contract #NAS5-32430 Goddard, "A Novel Negative Hydride Electrode for Ni-Metal Hydride Batteries", Phase I, Dec 92.

Exhibit 2. Quartz Chamber Arc Reactor Schematic (not to scale).